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Gas-Collection Technique for
Measurement of Fission-Gas Retention
in Irradiated Mo-UO₂ Cermets

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

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Clearinghouse for Federal Scientific and Technical Information
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Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

CESTI PRICES

H.Q. \$ 3.00; MW 65

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Report written: May 17, 1967

Report distributed: July 12, 1967

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A. J. Patrick

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GAS-COLLECTION TECHNIQUE FOR MEASUREMENT OF FISSION-GAS

RETENTION IN IRRADIATED Mo UO₂ CERMETS

by

A. J. Patrick

ABSTRACT

A method is described for determining the quantity of rare gas fission products retained in irradiated molybdenum-uranium dioxide cermet fuel specimens. Information concerning the location of the gas in the cermet structure is also obtained. The method utilizes differential dissolution of molybdenum from the cermet followed by the separate dissolution of the uranium dioxide. Cold-trap pumping of the gases liberated during boiling of the solutions enables a high percentage of the rare gases originally in the cermet to be collected in a sample can. Contents of the sample can are determined by mass spectrometric and gamma-ray counting techniques.

Introduction

In order to measure the fission-gas retention in Mo 40 v/o UO₂ cermets after long-term neutron irradiation tests, a technique was developed that used the differential dissolution of Mo and UO₂ and the measurement of the gas released when each component of the cermet dissolved. The Mo in the cermet was removed by electrolytic dissolution in 10 N NaOH, and then the solution was boiled to release Kr and Xe. (A cold-trap pumping arrangement swept these gases to a collection can.) During the electrolytic dissolution process, the UO₂ in the cermet settled to the bottom of the flask in particulate form. These particles were recovered, and a similar dissolution and gas-collection procedure was carried out with 50% HCL + 50% HNO₃ as the solvent.

No chemical reaction was indicated between the UO₂ and the solvent during Mo dissolution. This was apparent from the electron microscopic and x-ray

diffraction measurements of the UO₂ particles recovered from the cermet following the removal of the Mo, and it was also apparent because the Mo could be removed slowly enough to leave the UO₂ phase of the cermet physically intact; i.e., in a weakly interconnected spongelike structure.

The taking of repetitive samples for a given dissolution indicated that greater than 95% of the Kr and Xe was collected in one recovery operation for the Mo dissolution; gas-recovery percentages were more variable for UO₂ dissolution, although they were generally above 80%. Because of this variability, at least two sequential gas samples for each UO₂ dissolution were collected. The quantity of gas recovered was measured by counting ⁸⁵Kr and occasionally by the mass spectrometric analysis of the contents of the can. Retention fractions were calculated from radiochemical fission density

Fig. 1. Gas-collection system and associated equipment.

Fig. 2. Dissolving flask showing electrodes with fuel sample in dissolving position.

measurements based on the amount of ^{144}Ce and ^{95}Zr in the cermet. These retention fractions were checked against retention fractions observed for the cermet as a whole and were added to gas-release

Fig. 3. Liquid trap (to prevent carryover of liquid outside of hot cell).

measurements made by puncturing the irradiation capsule. In general, the measurements of recovered gas and the calculations of total gas produced agreed to within 10%.

The components of the gas-recovery system designed for hot-cell operation are shown in Figs. 1 through 5: Fig. 1 shows the vacuum system, Fig. 2 the dissolving flask, Fig. 3 a liquid trap (a safety feature), Fig. 4 the gas sample can, and Fig. 5 the electrical power network.

Procedural Details of Dissolution and Gas Recovery

An alligator clip attached to the positive electrode of the dissolving flask holds a section of an irradiated W-coated Mo UO_2 cylinder. (As illustrated in Fig. 2, two electrodes enter the dissolving flask through a rubber stopper, held in place by a clamp arrangement.) Twenty cubic centimeters of 10 N NaOH, placed in the dissolving flask, is frozen by a Dry Ice and methylene chloride bath. After the rubber stopper and electrodes are inserted, a vacuum of 5×10^{-4} torr is pulled on the

Fig. 4. Gas sample can.

dissolving flask. The flask is then closed off and the Dry Ice bath removed. After the NaOH thaws the fuel slug is immersed in the liquid to the required depth. The power leads (Fig. 5) are then attached to the electrodes, and a current of 5 to 7 amp is applied. This dissolves the W and Mo while precipitating the UO_2 .

A gas sample can (Fig. 4) with 2 g of sponge Ti placed inside to absorb the H liberated during dissolving is attached and heated to approximately 600°C . The dissolving flask is then heated until the liquid is at a slow boil. It is then open to the sample can. After 10 min of slow boiling, the sample can is cooled and placed in a liquid N_2 bath for 1 hr.

The liquid from the dissolving flask is then filtered through a glass filter paper, and the UO_2 particles are washed several times until they are

Fig. 5. Hot-cell wiring (schematic).

free of NaOH. The filter paper and UO_2 particles are dried.

The next step is to place 20 cc of 50% HCl + 50% HNO_3 in the dissolving flask. This is frozen by using a Dry Ice bath of methylene chloride. When the solution is frozen solid, the filter paper containing the UO_2 particles is placed in the dissolving flask and the flask is sealed. A vacuum (approximately 5×10^{-4} torr) is pulled on the system and flask. The flask is then closed off and the ice bath removed. The liquid thaws and starts dissolving the UO_2 particles; in approximately 4 hr the reaction between the acid and the UO_2 ceases and the gas released from the UO_2 particles can be collected.

A sample can is attached and pumped down to a vacuum of approximately 10^{-6} torr. The sample can is placed in a liquid N_2 bath and allowed to cool. The dissolving flask is then opened to the can and the liquid in the flask is heated to a slow boil for approximately 20 min. When heating is terminated, vapor from the liquid continues to be pumped into the cold sample can. This process is allowed to continue for another 40 min after which the sample can is removed from the system and the quantity of fission gas it contains is determined by gamma-ray counting and mass spectrometer measurements.

Conclusion

The fission-gas collection technique described above has been used to determine the quantity of fission gas retained in at least fifty samples of irradiated Mo UO₂ cermet. Measurements have been compared to results obtained when the procedure described above for determining the gas retention of the UO₂ fraction of the cermet is applied to the cermet as a whole, i.e., when the procedure for the separate dissolution of Mo is bypassed. Measurements of the amount of fission gas escaping from the cermet during irradiation have also been made. Some of these results are reported in Ref. 1. When the sum of the measured quantities of fission gas released and retained is compared with the quantity produced - as determined by radiochemical determination of total fissions - the two values show agreement within a deviation of 12%.

Acknowledgments

The author is grateful to W. A. Ranken for his encouragement and helpful suggestions and discussions. The author gratefully acknowledges the contribution of the following individuals and groups at Los Alamos Scientific Laboratory: C. E. Nilsson, engineering design of hot-cell equipment; M. C. Chaney, gas counting technique; Group CMB-14, hot-cell technique and facilities; and F. H. Newbury for hot-cell operation.

Reference

1. W. A. Ranken, "Second Status Report on High Temperature Irradiation of Mo UO₂ Fuel," LA-3462-MS, Los Alamos Scientific Laboratory, April 1966 (Confidential RD).